On the Rate of Decline of Persistent Organic Contaminants in Lake Trout (*Salvalinus namaycush*) from the Great Lakes, 1970-2003

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Introduction

The decline of banned persistent, bioaccumulative, and toxic contaminants (PBTs) in the environment is of keen interest to assess the success of the regulatory action as well as to estimate present and future risk of exposure to PBTs. Two PBTs of particular concern are polychlorinated biphenyls (PCBs) and dichloro-diphenyl-trichlorethane (DDT). Due to concerns of their presence in fish, birds, and wildlife in the Laurentian Great Lakes, and their demonstrated adverse effects in humans and animals, they were banned in North America in the mid-1970s by both Canada and the United States. However, these industrial chemicals are still present in top predators at concentrations that are sufficiently high as to trigger fish consumption advisories and close commercial fisheries.

Fish have often been the biomonitor of choice for assessing hydrophobic contaminants in the environment, as the analytical measurements for fish are far easier than they are for water samples, and they are a good indicator of ecosystem health. Fish monitoring programs of various levels of effort have been conducted in the Great Lakes since the 1970's by a number of state, provincial, and federal agencies, but have not had consistent methodologies or aims.

One exception is the Great Lakes Fish Monitoring Program (GLFMP), it was implemented as a coordinated effort that consisted of a Cooperative Agreement between the US Environmental Protection Agency (USEPA) Great Lakes National Program Office (GLNPO), the US Fish & Wildlife Service (and now the National Biological Division of the US Geological Survey, USGS-BRD) and the Great Lakes states *(1)*. However, in 2003 the USGS-BRD ended its relationship with EPA and the GLFMP making EPA the sole operator of the program. A similar and complimentary program, the Great Lakes Fish Contaminant Surveillance Program (GLFCSP), has also monitored organic contaminants in fish over time. The GLFCSP was operated by the Department of Fisheries and Oceans of Canada between 1977 and 2006 at which time the program was reorganized to be operated by Environment Canada. The primary goal of the GLFMP is to monitor time trends of bioaccumulative organic chemicals in the Great Lakes using lake trout (*Salvalinus namaycush*) in four of the lakes and walleye (*Stizostedion vitreum*) in Lake Erie. The primary data quality objective is to detect contaminant concentration changes of 25% with 95% confidence.

This successful program has provided one of the most useful long-term data sets of organic contaminants on record. Numerous reports and interpretations of these data have since been published *(2-10)*. These publications have shown that the concentrations of chemicals whose sources have been banned or restricted have declined in direct response to regulatory actions. Declines are usually described by simple first order exponential decay models. In addition, these data have been used to demonstrate that some contaminants such as PCBs may have stopped declining *(8,9,11)*, not because of new sources but because of other factors, including changes in the food web *(8,12,13)*.

In this report we will present and interpret the time trends for PCBs, DDTs, toxaphene, chlordanes, nonaclors, mirex, and polybrominated diphenyl ethers (PBDEs) in lake trout and walleye using GLFMP data from 1970 to 2003. The declines of banned or restricted chemicals in fish are corroborated here; however, these long-term data demonstrate that the observed first-order rate constants of most chemicals have significantly decreased in magnitude in most of the Great Lakes. These rate constants are very much influenced by the processing of PBTs within the lakes, and by the relative importance of atmospheric and sedimentary sources.

Methods

Fish Samples. All fish from this study were collected from two master spawning sites from each lake by trawl by the USGS-BRD as part of their stock assessment surveys. Prior to 1984, the even year sites were sampled annually while the odd year sites were more sporadic. After 1982, each site was sampled in alternate years (see Figure 1). Lake trout between 600-700 mm are collected from all lakes except Lake Erie; walleye between 400-500 mm are the top predator species selected for Lake Erie. Fish from a given site/lake were ground and homogenized into 10 composites of 5 whole fish each, frozen, and shipped to the analytical lab.

Sample Extraction and Interference Removal for Organic Compounds. The analysis of the lake trout/walleye composites for the GLFMP has been done historically by EPA's Contract Regional Laboratory (1970s), the Wisconsin State Laboratory of Hygiene (1980s), and the US BRD Analytical Laboratory (1990s). From 1999-2003, the analyses have been done under the direction of Dr. Deborah Swackhamer at the University of Minnesota, Environmental Chemistry Laboratories, School of Public Health. An extensive and rigorous Quality Assurance Program has been adhered to by all laboratories in order to ensure the comparability of data across the different laboratories, and to ensure high quality data from any given laboratory.

Methods prior to 1999 have been summarized elsewhere *(8)*. The methods for samples from 1999-2003 are summarized below; detailed methods can be found in the EPA-approved Quality Assurance Project Plan *(14)*. Homogenized subsamples of ground fish are extracted by Soxhlet extraction for 4 hours with methanol followed by 18 hours with dichloromethane. Surrogate standards are added to every sample at the beginning of extraction. A subsample is removed for lipid analysis. The fraction lipid is determined gravimetrically by drying the subsample to constant dry weight.

Interferences are removed by liquid-solid chromatography using 6% deactivated alumina and eluting with hexane. This extract is then placed on a column containing fully activated silica gel over 1% deactivated neutral alumina (w/w), with anhydrous sodium sulfate above and below each layer. The column is eluted with hexane (Fraction 1). The column is further eluted with 40%/60% DCM/hexane (Fraction 2). Prior to instrumental analysis, the extract is reduced in volume and the internal standard is added (PCB #204) to Fraction 1.

Extract Instrumental Analysis. The analysis for PCBs is done on Fraction 1. Once the data have been reviewed and found to be acceptable, then Fraction 1 and 2 are recombined. This is because of the high potential of interference from other compounds in PCB analyses. In our experience, recoveries of other analytes are improved if the two fractions are recombined. PCB congeners (110 congeners or congener groups) are analyzed by gas chromatography with electron capture detection (GC-ECD) using similar methods to previous studies from this laboratory *(15-18)*.

All other organochlorines, including DDT, DDE, and DDD are analyzed by gas chromatographic mass spectrometry with electron capture negative ion detection (GCMS-ECNI). Selected ions are monitored for quantifying and confirming each compound.

Summary Statistics. Arithmetic means and 95% confidence limits are computed for the lake trout or walleye for each lake and each year. Samples of the same species from the same lake and year are considered field replicates. In general, values with a QA qualifier are not included; however, all data and statistics are reviewed by the PI and professional judgment is used to determine which values are included in the summary statistics. Total PCB is calculated as the sum of all 110 PCB congeners; $\sum$ DDT is the sum of p,p-DDD, p,p-DDE, and p,p-DDT (hereafter referred to as DDT).

Quality Assurance. Duplicate analyses of a single composite are used to assess analytical precision. Precision for PCBs and DDT is generally within 10%. Overall program precision is determined from the standard deviation of the mean of the 10 composites for each site and year. Generally overall precision was within 50%. Surrogate recovery standards are used to assess accuracy, as well as analyses of a GLFMP-generated "check fish" samples. Recoveries of all surrogates were > 50%. All data were corrected to surrogate recovery except for PBDEs. Values for the "check sample" were ± 20% of the reference values from USGS-BRD. All procedural blanks were below detection. Method detection limits (MDLs) for 99% confidence were determined as mass/extract and defined as 3 standard deviations of 7 runs of a blank spiked with a very low level of analyte (40 CFR Part 136, Appendix B, Rev.1.11, October 26, 1984). See also the EPA-approved Quality Assurance Project Plan for greater detail *(14)*.

Results and Discussion

Lake Trout Data 2001-2003: overall observations

The mean values of all analytes for each year (2001, 2002, 2003) are provided in Table 1. Concentrations are for whole lake trout and reported as nanograms of contaminant per gram fish on a wet weight basis. All individual concentrations were corrected to the appropriate surrogate recovery, and reviewed carefully for compliance with the QA objectives and guidelines. Mean values only include those data that passed QA review. All individual data as well as means are reported in the Great Lakes Environmental Database (GLENDA), http://www.epa.gov/greatlakes/monitoring/data_proj/glenda/index.html.

The following contaminants were found routinely in lake trout from lakes Ontario, Huron, Michigan, and Superior: PCBs, p,p'-DDE, p,p'-DDT, HCB, a-HCH, dieldrin, octachlorostyrene (except Superior), toxaphene, cis- and trans-chlordane, cis- and trans-nonachlor, PBDE congeners, and mercury. The walleye from Lake Erie had these contaminants with the exception of p,p'-DDT, and a-HCH, which were below method detection limits. Mirex was found routinely in lake trout from Lake Ontario, but was not detected in lake trout or walleye from the other lakes. Heptachlor epoxide was detected only in lake trout from Lake Michigan.

The GLFMP is designed to track trends in contaminant concentrations within a lake. Fish are collected within a narrow length range. As the length-age relationship varies between lakes, it is not possible to directly compare concentrations across lakes. In addition, walleye, rather than lake trout, are collected from Lake Erie. However, some general conclusions can be drawn. Concentrations of ∑PCB, ΣDDT, and other contaminants are generally lowest in Lake Superior and greatest in Lakes Michigan and Ontario. This general pattern is consistent with that observed in other programs and media. The same general pattern was observed in fall run coho salmon *(7)* and in the water column *(19)*. Exceptions to this are alpha-HCH and toxaphene which are greatest in Lake Superior *(20-22)* and OCS and mirex which are greatest in Lake Ontario.

General Summary of Contaminant Trends in Lake Trout

The data for all contaminants routinely detected by the program are shown in Figure 2; means for each year (1970-2003) for all contaminants can be found in Table 1. All individual data are reported in the Great Lakes Environmental Database (GLENDA), http://www.epa.gov/greatlakes/monitoring/data_proj/glenda/index.html) This work will focus on data from the sites with the longest sampling records in each lake, now sampled in even years. In general, concentrations of DDT, PCBs, dieldrin and oxychlordane have all declined significantly in Great Lakes fish since the monitoring of these chemicals began in the 1970s. For example, in Lake Michigan, DDT has declined by 96% since 1970; since peaking, PCBs, dieldrin, and oxychlordane have declined by 90%, 78%, and 83%, respectively. *Trans*- and *cis*-nonachlor, *cis*-chlordane, and toxaphene, which were not monitored until the mid-80s, generally showed decreases in concentration as well; dramatic decreases of mirex have occurred recently in Lake Ontario. Monitoring of PBDEs began in 2000, and our data combined with data obtained from archived samples *(23,24)* show initially rapidly increasing concentrations that now are beginning to level off.

Data for additional chemicals added to the program in the 1980s or 90s can be found in Tables S1 and S2; discussions of these contaminants in Great Lakes fish can be found elsewhere *(8-10)*. Overall, the Great Lakes Fish Monitoring Program has demonstrated that the regulatory programs introduced in the 1970s were initially effective in reducing contamination levels in the Great Lakes for the targeted chemicals.

Previous studies have modeled the decrease in concentration of" legacy" (restricted or banned) contaminants assuming first order decay, resulting in reasonably good R^2 values (except for Lake Superior lake trout)*(8)*. As an example, Figure 3 shows concentrations of DDT in Lake Michigan lake trout over time with an exponential model fit (red dashed line). The R^2 value is 0.95; however, the semi-logarithmic plot on the bottom of Figure 3 clearly shows the data do not follow exponential decay over the entire monitoring period. If they had, we would have expected DDT concentrations to be about an order of magnitude lower than what is observed currently.

The solid blue line on Figure 3 shows another model, one that assumes exponential first order decay to a non-zero constant. There is little improvement in the R^2 value (0.96); however, the qualitative fit is clearly superior. The earlier, robust decrease in DDT in Lake Michigan has slowed substantially, if not halted altogether. It has been noted previously that the decline of many contaminants in the Great Lakes does not always fit a simple first-order exponential decay model over the entire dataset *(8,9,25-30)*. The most recent data confirm these observations.

An Analysis of the Change in Rate Constants with Time

The apparent change in rate constants in the mid-1980s led us to conduct a more thorough analysis using rate constants calculated from subsets of the data. There is a trade-off between resolution and precision in choosing the size of the subset; we chose to use six consecutive data points as a balance between these two trade-offs. Rate constants were calculated assuming first-order behavior over the short amounts of time involved (5-10 years). The results for five contaminants (PCBs, DDT, dieldrin, oxychlordane, PBDEs) are shown in Figures 4 and 5.

In general, we see the same pattern for lakes Superior, Huron, and Michigan and for three of the four legacy contaminants (DDT, PCB, and oxychlordane) that have been monitored since the 1970s. After peak concentrations for dieldrin and PCBs in Lake Michigan, or at the inception of monitoring for other lakes, rate constants were negative, finding a minimum in the early 1980s. Half-lives of the chemicals in the "early period" (mid-1970s – mid-1980s) were typically 3-6 years. In the mid-1980s the rate constants became less negative and approached zero more recently. Half-lives increased to 15-30 years or became infinite. Lake Superior generally has greater rate constants than the other lakes for the later time period rate constants. On a year-to-year basis, there appears to be greater consistency within a lake for multiple contaminants (Figure 5) than there is for a given contaminant across the different lakes (Figure 4). This behavior underscores the importance of fate and transport processes on contaminant behavior within a given lake and the fact that these processes differ among the lakes.

In Lake Ontario, trend monitoring did not begin until 1982, due to lack of suitably-sized lake trout; from that point on, the data show similar trends as in the upper Great Lakes. Lake Erie data are for walleye instead of lake trout, making inter-lake comparisons difficult. Also, Lake Erie is physically very different from the other Great Lakes. It is quite shallow compared to the rest of the lakes, with an average depth of 19 meters, compared to 147, 85, 59, and 86 meters for Lakes Superior, Michigan, Huron, and Ontario respectively *(31)*. The sediment-water interactions of Lake Erie may confound comparisons of it to the other lakes.

The manufacture of one group of contaminants, PBDEs, has only recently been curtailed, and thus they do not have the same trends as other legacy contaminants (Figure 4). In general, the rate constants for PBDEs in lake trout are positive, but they have been decreasing over the past several decades, currently nearing values of zero. This means PBDE concentrations in lake trout may be close to stabilizing, and, if inputs do not increase or are further reduced, could soon begin to decrease. Similar trends have been seen in herring gull eggs *(32)*. Lake Superior appears to have a fairly stable rate constant over the time period of measurement, although the error associated with these short-term rate constants is large.

Rates of change for mirex in Lake Ontario lake trout also change over time. Half-lives calculated from published lake trout data for lake trout collected prior to 1995 *(33)* are around 10-20 years. Both GLFMP sites, however, have half-lives of 2-3 years for post-1995 data. Similarly, mirex concentrations were stable in Lake Ontario salmon until 1992, but decreased dramatically between 1992 and 1999 *(34)*. Makarewicz et al. suggest the most important contribution to this change is remediation activities in the mid-1990s at the Hooker Chemical Company Niagara Falls site, a known source of mirex, but caution further data are needed for confirmation *(34)*. The lake trout data presented here from 1999-2003 provide further confirmation of this trend.

Early and Late Period Rate Constants

Although there is considerable scatter in the data, the rate constants of PCBs, DDT, and oxychlordane in the upper lakes appear to have two relatively stable periods: post-peak to the mid- to late-1980s, and the most recent data from the 1990s forward. This would be consistent with several previously proposed models that have a fast, early decay followed by stable concentrations or a slower, secondary decay *(25,27,29)*. Previous work has attempted to use modeling to find the date of the rate change, or change point *(8)*. Because of scatter in the data, however, we cannot pinpoint a precise date when the rate constants change (and, if the models are correct, it is a gradual change), but we can identify a reasonable range in dates when the change in apparent first-order rate constants has occurred. This range of dates seems to occur at slightly different times for DDT, PCBs, and oxychlordane, with the order being PCBs first (approximately 1986), then ΣDDT (approximately 1990), then oxychlordane (approximately 1992). PCB production was banned in the United States in 1977, DDT was banned from use in the U.S. in 1973 and Chlordane (oxychlordane is a component of the technical mixture) was banned from commercial production and sale in the U.S. in 1988.

If we divide the data into two periods based on when the change in rate constants occurred we can calculate apparent first-order decay rate constants with smaller errors than the rate constants shown in Figures 4 and 5. Table 2 shows the results of these calculations for most contaminants with five or more detections in all lakes except Lake Erie. Because of the uncertainty in the date and nature of the rate constant change, the early rate constants are calculated using three consecutive ending dates, and the later rate constants are calculated using three consecutive starting dates, whenever possible. The starting and ending dates were chosen from visual inspection of Figure 4. Lake Michigan early rate constants for PCBs are also calculated using three consecutive starting dates to estimate the center on the observed peak concentration. Rate constants for dieldrin are calculated starting from the date of the peak or adjacent dates and continuing through 2002 or 2003. The corresponding half-lives of select contaminants have been summarized in Table 3.

The regressions from the earlier time periods in the upper lakes show significant negative rate constants and short half-lives. The rate constants do not appear to depend on starting or ending dates for the regressions, varying in a narrow range with half-lives of about 3-6 years for PCBs, DDT, and oxychlordane in all three lakes. Later rate constants in the upper lakes show much slower decay for PCBs, DDT, and oxychlordane, and much longer half-lives, on the order of 15 yrs or greater. In most cases, the decline in later years is not statistically significant. Rate constants for this later period can also be calculated for the sites sampled in odd years (Table 2), and are generally consistent with the even-year rates, with the exception of Lake Superior. Nearly all calculated rate constants for the even-year Lake Superior site over this time period indicate increasing concentrations, although in only one case are these positive rate constants significantly different from zero. Concentration data from lake trout collected at the odd-year Lake Superior site, Keweenaw Point, do *not* show an increasing trend (Table 2; see further discussion below).

Dieldrin does not follow this pattern, likely because its use continued into the 1980s and because it is also a degradation product of aldrin. A similar observation has been made with herring gull data *(26)*. Following the peak in the late 1970s, rate constants have remained steady. Dieldrin has half-lives of about 10 years in Lakes Huron, Ontario, and Michigan over the entire monitoring period since the peak concentration. In Lake Superior, the half-lives are much greater, nearly 25 years. All rate constants calculated for dieldrin are significantly different from zero among the sites that have been sampled since the 1970s.

The pattern of rate constants can be seen more clearly in Figure 6. Figure 6 also shows early and late rate constants for dieldrin, using the same dividing point as DDT, for illustration, and later rate constants for *trans*- and *cis*-nonachlor, *cis*-chlordane, and toxaphene, for which earlier data are not available.

Possible Mechanisms of Change in Rate Constants and Half-lives over Time

The apparent rate constants shown in Figure 6 show several types of behaviors: a fast initial decrease followed by rate constants near zero for DDT, PCBs, and oxychlordane; an unchanging rate constant for dieldrin; and a consistent pattern among chemicals and lakes for the slower, later rate constants. These observations are likely to be the result of a composite of different mechanisms changing over time in the Great Lakes ecosystems.

The fast initial decrease in concentration followed by a slower decrease or stabilization has been observed in other Great Lakes media besides fish. While the amount of data available for other media is not sufficient to conduct as thorough an analysis, rate constants for PCBs in air *(35)*, water *(36)*, and herring gull eggs *(37)* in the Great Lakes region were generally more negative in the late 1970s and early 1980s than they have been more recently (Figure 7). The most dramatic differences are seen in water. In sediments, there may be little difference in rate constants between time periods *(38)*, although more recently, there is some indication that sediment PCB concentrations may have stabilized in some lakes *(39-41)*.

What are the possible explanations for this general pattern of rapid declines in the 1970s and a leveling off of concentrations in the late 1980s - early 1990s? This is a change in the *rate* of change, over decadal time periods, which occurs for several banned, persistent contaminants in the three upper Great Lakes. Factors that could affect the rate of change include: (1) changes in the lake trout food web; (2) changes in fishery dynamics, including fish condition factor, age-weight relationships, size class distribution, etc; (3) climate change; and (4) changes in the source functions of the contaminants to the lakes.

Changes in the lake trout's food web have been observed in other Canadian lakes following the introduction of new fish *(42)*, and observed changes in the herring gull trophic levels have been found to significantly affect calculated rate constants for contaminant loss at many sites *(43)*. However, the fast initial decrease followed by stabilization is seen for different contaminants, for different lakes, and in several different media. While the similar behavior of different contaminants could be explained by changes in the lake trout food web or by changes in their condition factor, such changes would not explain the pattern seen in water and air. It would also be unlikely that these changes would occur at similar times in the biological communities of multiple lakes. Finally, in order to account for the deviation from exponential decay seen in Figure 3, the length of the lake trout food web would have to be increasing by a relatively constant amount each year over a period of 15 years. This scenario seems unlikely, although undoubtedly changes in lake trout food webs over time can account for at least a portion of the variation observed, especially year to year variations *(9)*. The long-term observations reported here, however, are not readily explained by either possible changes in food webs or changes in the fish community structure or condition.

Climate changes have occurred over the entire Great Lakes region during the last several decades. Lake Superior water temperatures have increased more than 1 ^{o}C per decade since the mid-1980s; over the same time period, air temperature has warmed somewhat less, wind speed has increased, ice cover has decreased, and stratification has come earlier *(44)*. These factors, as well as precipitation, all directly affect atmosphere/lake contaminant transport. Additionally, atmospheric concentrations of PCBs are not only a function of local air and water temperatures but may also be linked to global-scale climatic phenomenon *(45)*. Finally, PCB concentrations in phytoplankton, the base of the food web, have been shown to be influenced by temperature *(16)*. Previous work has postulated that regional climate can affect contaminant uptake and transfer through the food chain in order to explain basin-wide similarities in deviations from first-order decay for contaminants in herring gull eggs *(46)*.

We would expect that climate change would impact contaminants having similar physical-chemical properties in a similar manner. This was not apparent in later analyses of the gull egg data *(26)*, but is consistent with the lake trout data for PCBs, DDT, and oxychlordane presented here. Dieldrin is different, but this can be partly explained by the fact that it is a degradation product of aldrin and thus has a different source function. Other contaminants do not have sufficient data to make an assessment. However, the two-step decline in the upper lakes is not consistent with climate change impacts, if source functions remain constant. An increase in the air-water interface temperature and wind speed and an extension of the stratified period should result in an increase in the volatilization rate from water and an increase in the flux of contaminants from the water column, if the net flux from the lake is positive. Increases in water temperature should also increase primary production and the delivery of contaminants to the sediments and decrease the accumulation in fish. In fact, the opposite has been observed.

The remaining mechanism of change, changes in the source functions (external inputs to the water column) of the contaminants, is known to have occurred. Consider a box model showing all the inputs, losses, and internal processes affecting contaminant concentrations over time (Figure 8). Point source discharges have been eliminated or are negligible, and the primary sources (and sinks) for the lakes for most contaminants are now the atmosphere and sediments. As air (vapor) and (dissolved) water concentrations approach equilibrium, the air-water exchange rate should decline, and this would result in changes to their rate constants as observed in Figure 6. Furthermore, as air-water exchange declines, the importance of sediment-water exchange increases, and sediment pore water inputs may become a non-trivial input to the water column. All of this is to say that the regional and global reservoirs of these persistent, hydrophobic contaminants will mediate and slow the loss rate as the various compartments get closer to steady state or equilibrium. Water concentrations are a function of these inputs and losses. As concentrations in fish reflect concentrations in water, the change in source functions could be the primary factor behind rate changes observed in fish.

Unlike the other changes discussed, changes in the source functions *are* consistent with the observed pattern of contaminant concentrations in Great Lakes media. The earlier rapid declines can be attributed by their timing to the cessation of intentional releases and releases during use following regulation. A similarly sudden change in inputs occurred in the case of mirex in Lake Ontario after remediation. Following such events, losses exceeded inputs in the Great Lakes, and concentrations dropped sharply. However, additional, smaller sources remain. These sources are concentrated in urban areas in the Great Lakes region where large contaminated sites and multiple poorly characterized (or unknown) and difficult-to-control sources of PCBs and other industrial chemicals result in elevated atmospheric concentrations and contaminated tributaries and harbors *(47,48)*.

These remaining sources would be relatively constant in magnitude, and, indeed, the behavior of legacy contaminants in the Great Lakes in recent years is consistent with a system near steady state, where sources are decreasing little and only slightly smaller in magnitude than sinks. The small decrease in sources can likely be attributed in part to remediation of large contaminated industrial sites, such as we observed with mirex. The other part of the decline would be due to depletion of the sources. This implies that without further significant remediation, future decreases in sources will be the result of source depletion only and will occur at an even slower pace than in recent years.

The concentrations of these legacy contaminants are stabilizing in the upper Great Lakes. However, the resulting "stable" concentrations are still sufficiently great so as to cause negative impacts, such as fish consumption advisories. It is difficult to separate out the effects of ecosystem and climatic changes in order to predict future rates of decay, but contaminant sources at least may decrease at an even slower rate in the future. It is likely that these contaminants in fish and water will have impacts for decades to come.

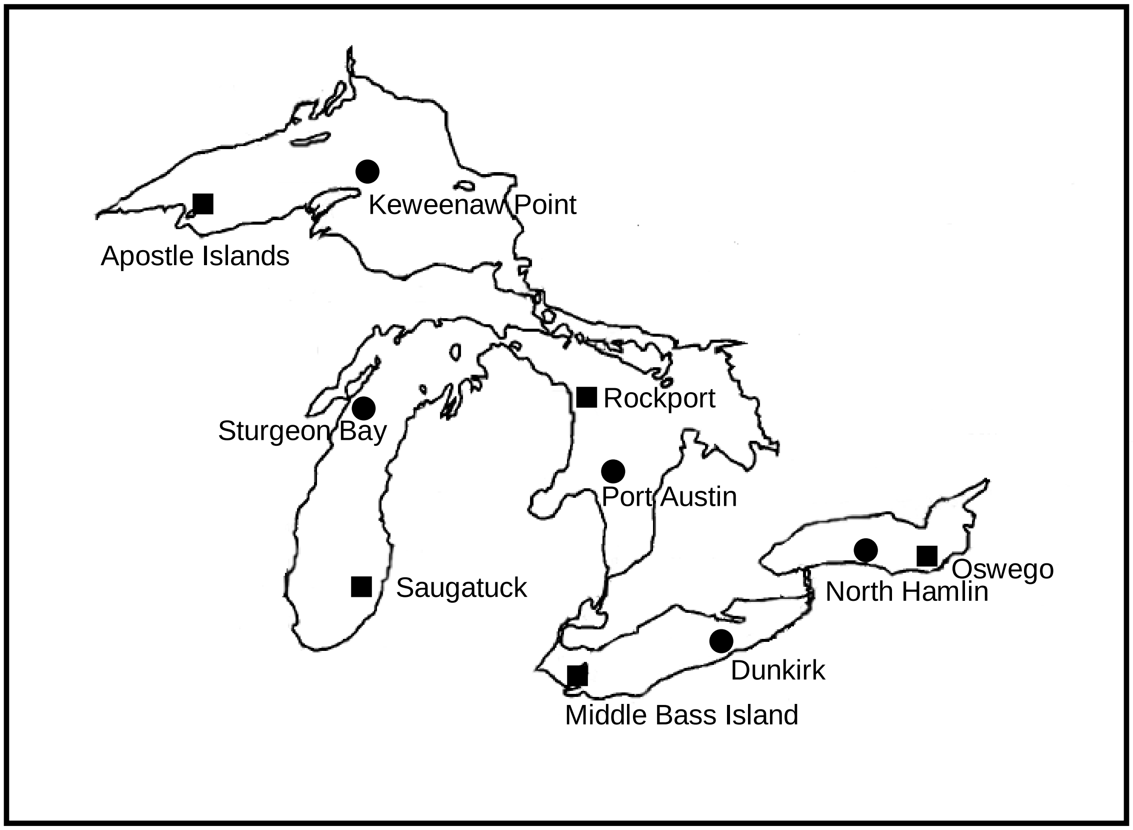

Figure 1. Sampling locations for lake trout and walleye. Points marked with a circle were sampled in odd years after 1983; those with squares were sampled in even years after 1983.

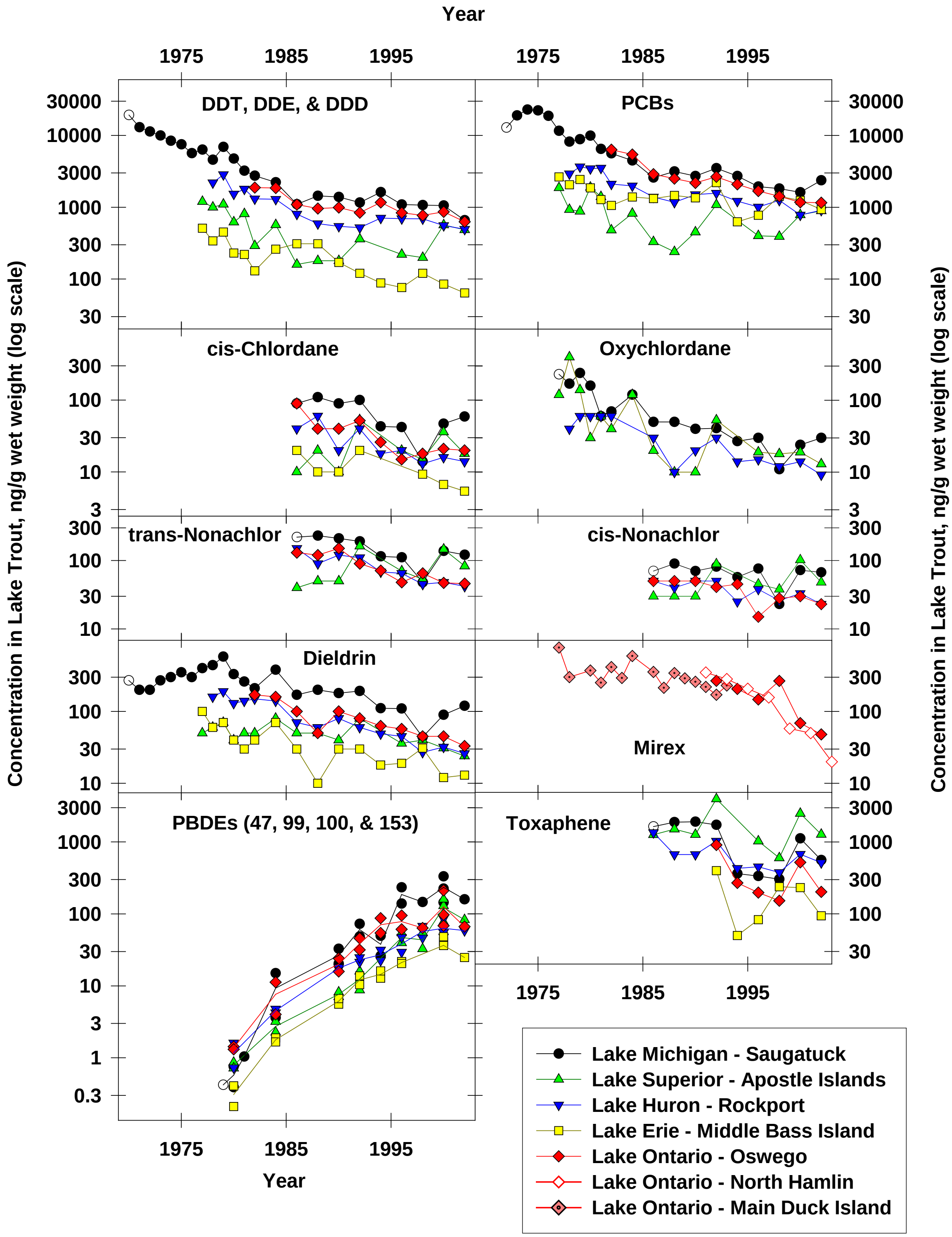

Figure 2. Concentrations of contaminants in lake trout and walleye (Lake Erie only). PBDE and mirex data are mainly from the literature *(23-24; 33)*; lines for PBDEs show, in some cases, averages of two data points for a given year. Mirex data from the Canadian fish program former master site at Main Duck Island in Lake Ontario are included for visual comparison but were not included in the statistical analyses.

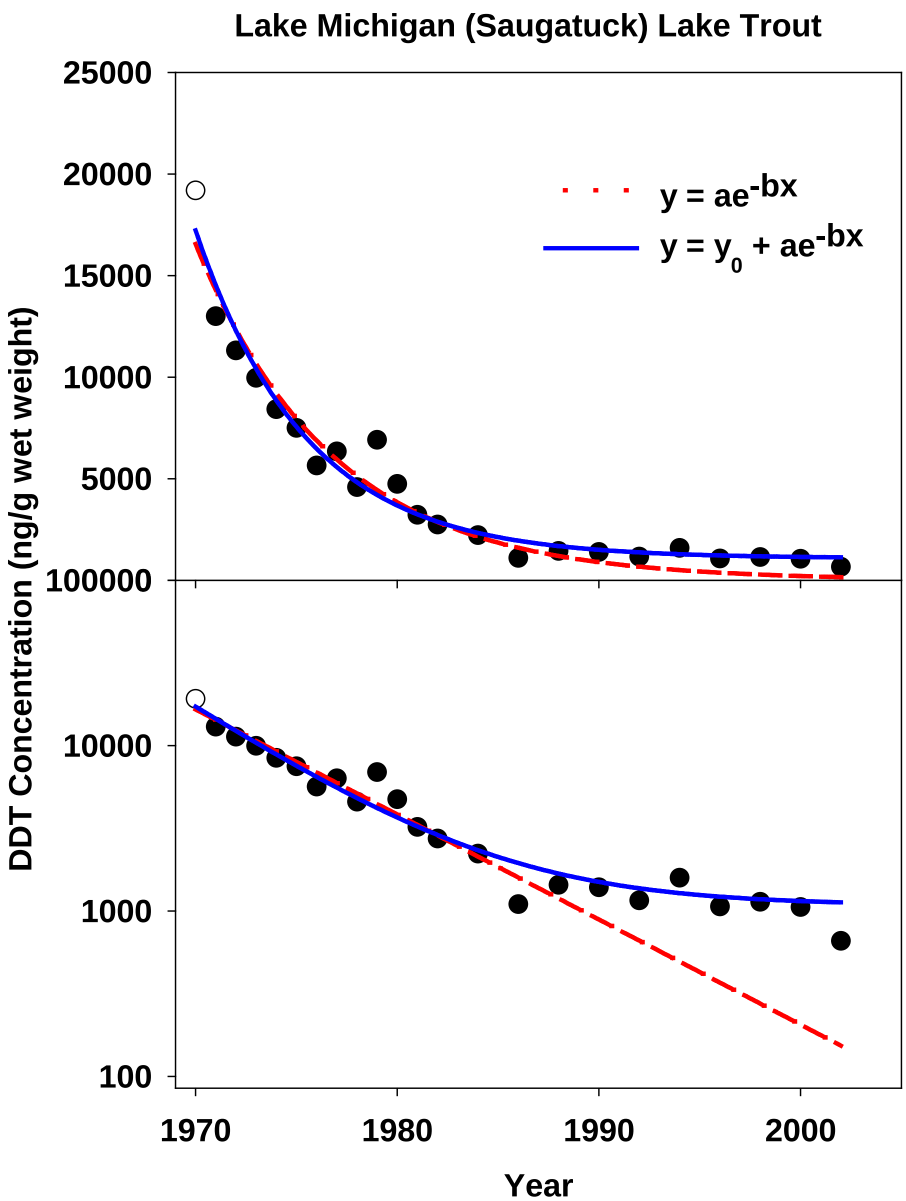

Figure 3. Concentrations of DDT in Lake Michigan lake trout using two models. Note the y-axis is logarithmic on the bottom portion of the figure.

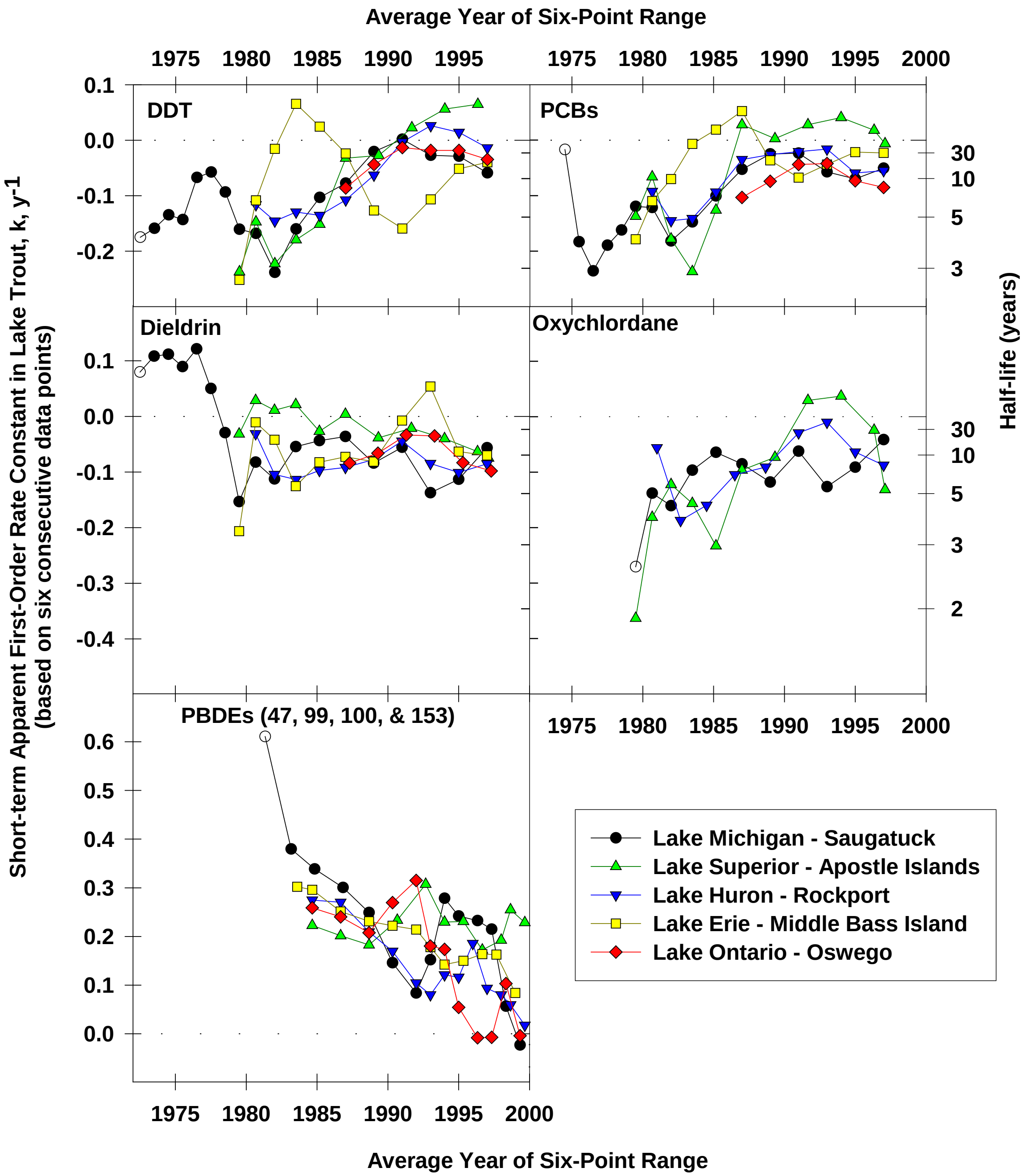

Figure 4. Calculated first-order rate constants for contaminants based on six consecutive data points, showing the change in the rate constants over time. Error bars are not shown for clarity, but are typically on the order of ±0.1 y^{-1}.

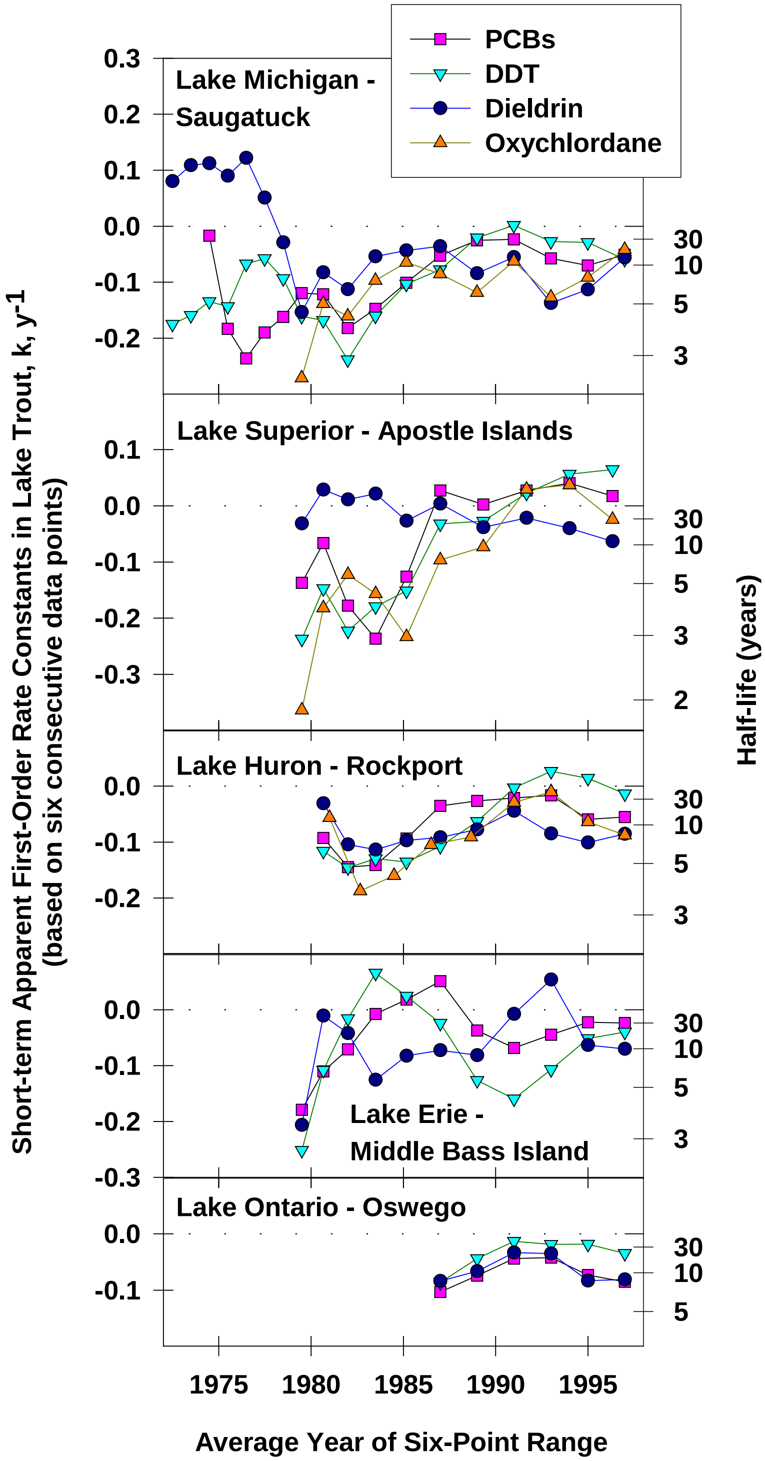

Figure 5. Calculated first-order rate constants for contaminants based on six consecutive data points, showing the change in the rate constants over time. Error bars are not shown for clarity, but are typically on the order of ±0.1 y^{-1}.

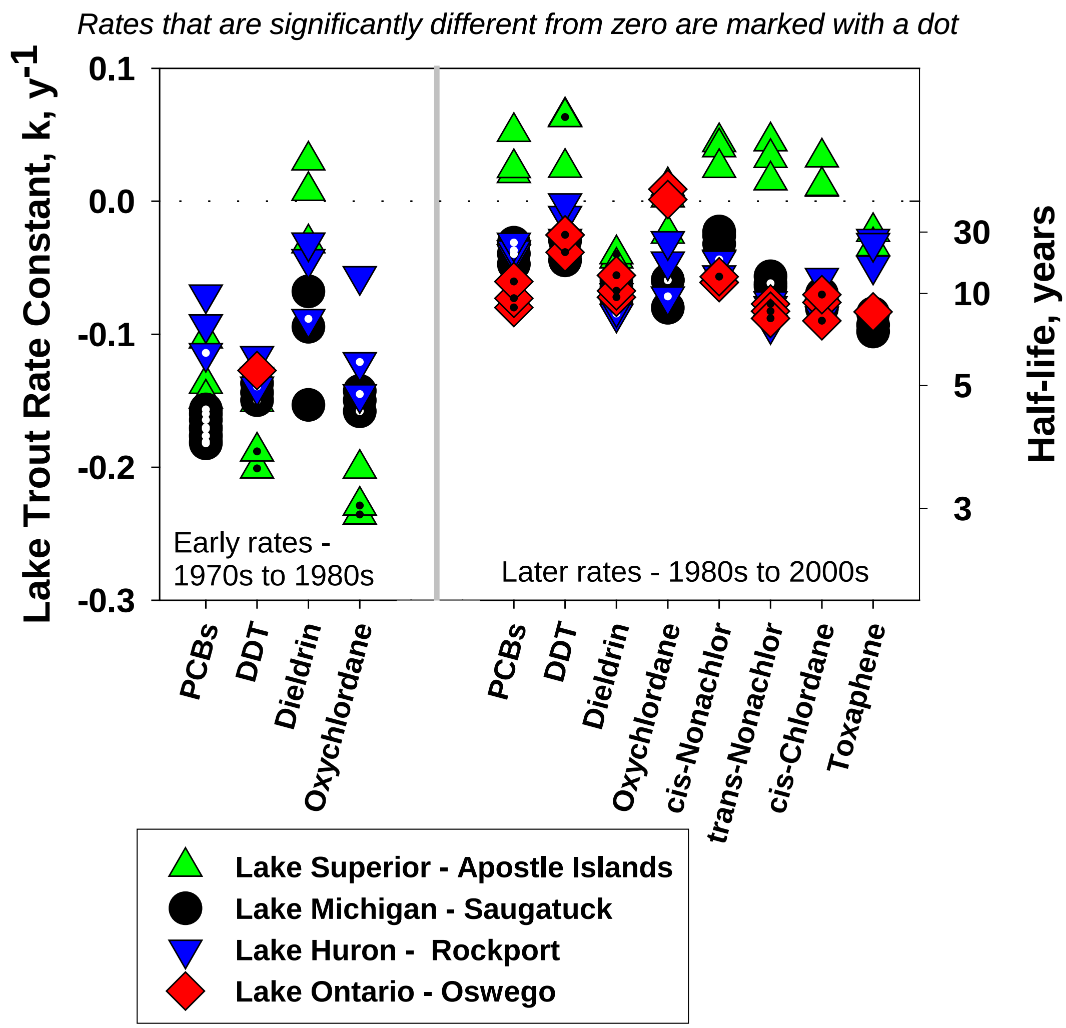

Figure 6. Rate constants calculated by time period; see Table 2 for details. Early and later rate constants for dieldrin are not shown in Table 2, but were calculated, as for PCBs, with starting and stopping dates in 1982, 1984, and 1986 for comparison with other contaminants.

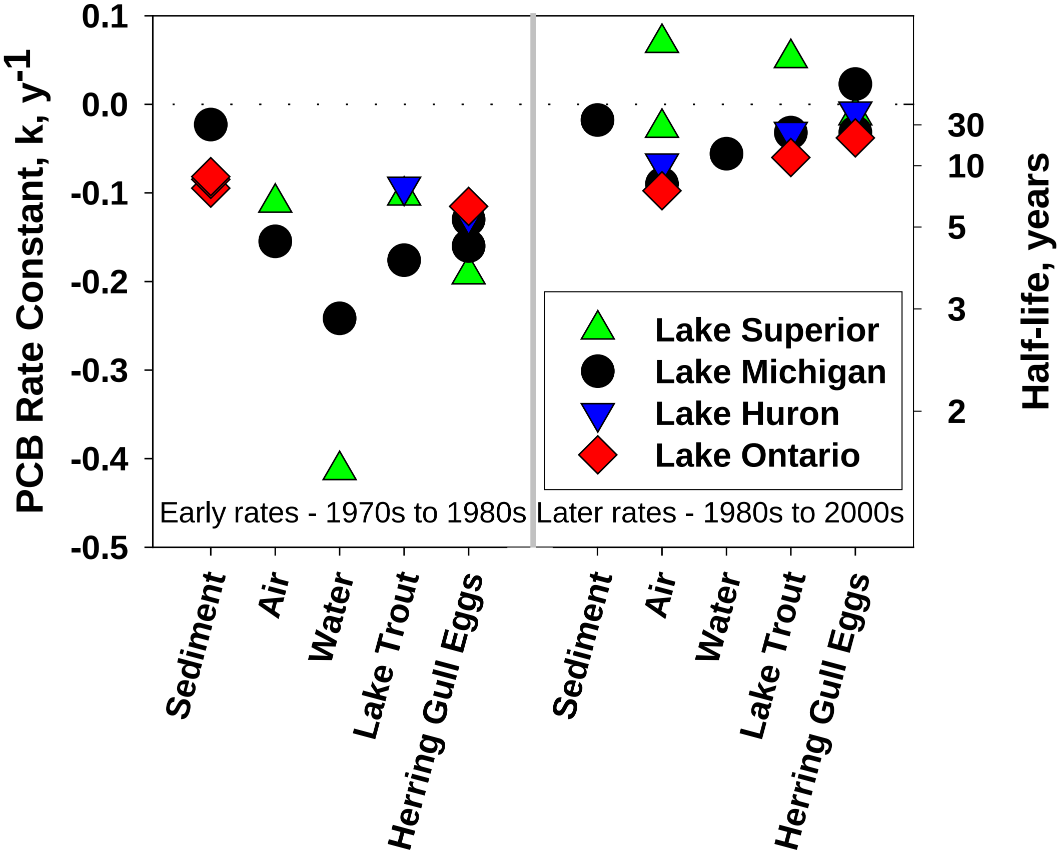

Figure 7. Rate constants calculated for PCBs in different Great Lakes media *(35-38,49)*. Dates of calculation vary based on availability of data, but whenever possible, the date of division is 1986. For herring gull data, the only sites included were those where adjusting for changes in trophic level did not change the rate constant by more than 25% when calculated for the entire data collection period *(43)*.

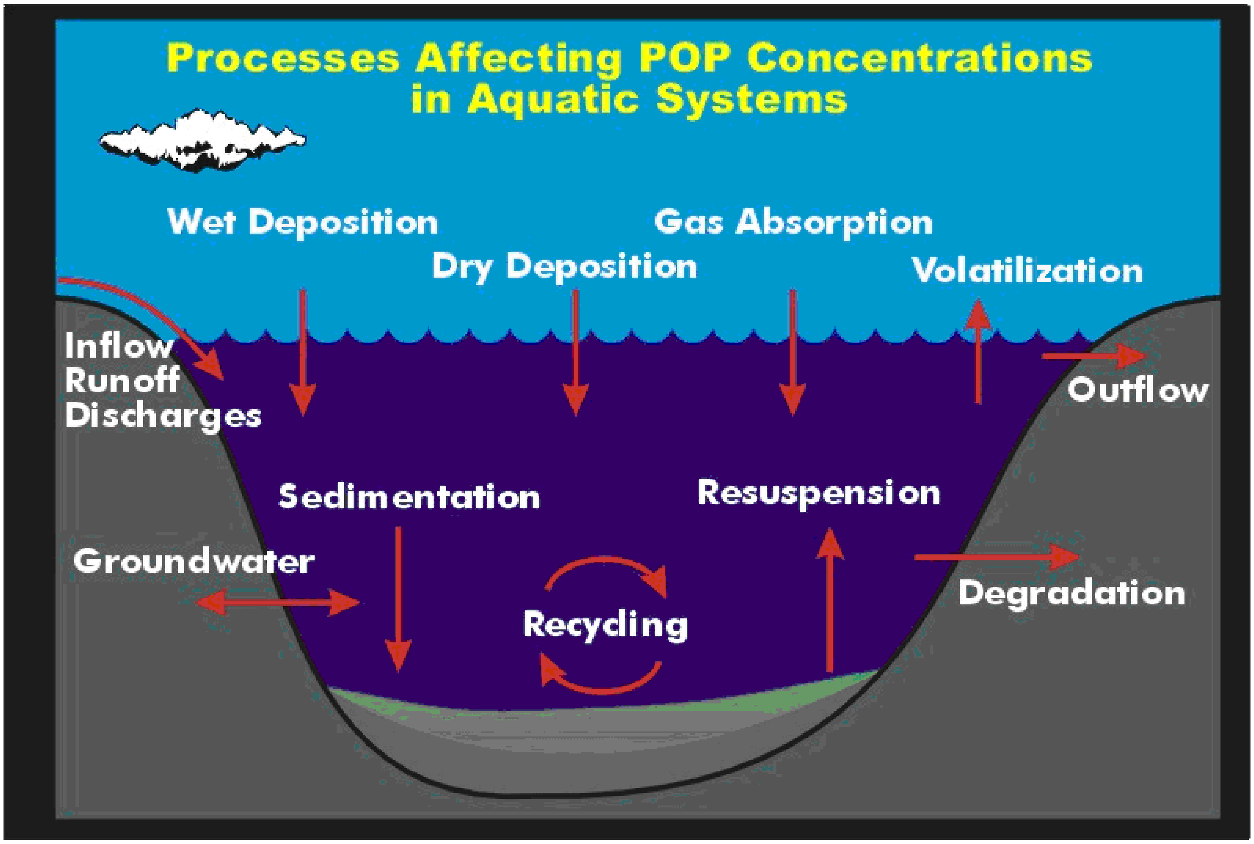

Figure 8. Air-water-sediment model system showing contaminant fluxes between compartments. Air-water exchange and sedimentation are the dominant fluxes for PCBs and other POPs in the Great Lakes. The system may be approaching steady state resulting in a stabilization of water concentrations at a non-zero concentration. Air concentrations may be affected by regional as well as global rates of decline.

Table 1. Mean concentrations and 95% confidence limits of target analytes in fish composites (lake trout and walleye) for 2001, 2002, and 2003. ND indicates the analyte was below the method detection limit.

Lake	Species	Yr	Statistic	Total PCBs	Dieldrin	HCB	OCS	alpha-HCH	Lindane	Heptachlor epoxide b
Superior	**LT**	**2001**	**mean**	**296**	**24**	**7.6**	**1.9**	**8.1**	**1.0**	**nd**
			95% CL	154	17	0.7	0.8	2.0		
			CV	46%	61%	9%	29%	21%		
	LT	**2002**	**mean**	**891**	**24**	**11**	**4.1**	**8.9**	**1.3**	**4.1**
			95% CL	117	5.5	0.91	0.66	1.5	0.19	
			CV	19%	35%	13%	25%	25%	15%	
	LT	**2003**	**mean**	**242**	**6.0**	**3.2**	**2.6**	**5.2**	**nd**	**nd**
			95% CL	41	1.6	0.41	0.97	0.69		
			CV	30%	23%	20%	59%	20%		
Huron	**LT**	**2001**	**mean**	**1059**	**43**	**10**	**5.6**	**1.6**	**nd**	**8.0**
			95% CL	218	17	2.3	2.0	0.3		
			CV	33%	59%	37%	59%	26%		
	LT	**2002**	**mean**	**908**	**26**	**8.9**	**2.3**	**1.3**	**0.34**	**nd**
			95% CL	51	2.7	0.42	0.14	0.1		
			CV	9%	17%	8%	10%	13%		
	LT	**2003**	**mean**	**845**	**55**	**4.9**	**5.6**	**1.3**	**nd**	**7.7**
			95% CL	81	51	0.53	0.65	0.26		
			CV	17%	124%	17%	19%	32%		
Michigan	**LT**	**2001**	**mean**	**1857**	**137**	**9.0**	**3.0**	**1.7**	**nd**	**16**
			95% CL	112	86	1.9	0.7	0.3		4.4
			CV	39%	72%	35%	40%	26%		38%
	LT	**2002**	**mean**	**2373**	**120**	**13**	**2.2**	**1.2**	**0.44**	**14**
			95% CL	690	53	1.9	0.33	0.31		5.8
			CV	36%	45%	15%	16%	27%		37%
	LT	**2003**	**mean**	**1263**	**88**	**5.0**	**2.3**	**1.1**	**nd**	**13.3**
			95% CL	172	44	0.32	0.46	0.11		2.2
			CV	24%	36%	9%	21%	15%		25%

Lake	Species	Yr	Statistic	Total PCBs	Dieldrin	HCB	OCS	alpha-HCH	Lindane	Heptachlor epoxide b
Erie	**Walleye**	**2001**	**mean**	**812**	**9.8**	**2.75**	**5.9**	**0.64**	**nd**	**nd**
			95% CL	85	2.2	0.7	1.7	0.1		
			CV	17%	26%	35%	45%	21%		
	Walleye	**2002**	**mean**	**934**	**13**	**2.9**	**4.4**	**1.5**	**nd**	**nd**
			95% CL	203	5.1	0.37	1.2	1.1		
			CV	29%	52%	19%	41%	110%		
	Walleye	**2003**	**mean**	**607**	**8.7**	**2.1**	**3.5**	**0.28**	**nd**	**nd**
			95% CL	122	3.2	0.12	0.99	0.082		
			CV	37%	45%	5%	46%	33%		
Ontario	**LT**	**2001**	**mean**	**996**	**36**	**16**	**17**	**2.1**	**1.1**	**nd**
			95% CL	185	16	3.1	3.6	0.3	0.3	
			CV	30%	51%	32%	34%	20%	19%	
	LT	**2002**	**mean**	**1157**	**33**	**18**	**8.7**	**13**	**1.1**	**nd**
			95% CL	202	11	2.9	1.9	11	0.16	
			CV	29%	48%	27%	35%	130%	13%	
	LT	**2003**	**mean**	**1092**	**137**	**7.8**	**21**	**1.4**	**nd**	**nd**
			95% CL	186	141	0.91	2.9	0.25		
			CV	30%	129%	19%	22%	26%		

Lake	Species	Yr	Statistic	Oxychlordane	Endrin	Mirex	p,p-DDE	p,p-DDD	p,p-DDT	Sum, p,p-DDE+DDT+DDD
Superior	**LT**	**2001**	**mean**	**nd**	**nd**	**nd**	**135**	**nd**	**36**	**160**
			95% CL				28			51
			CV				15%			23%
	LT	**2002**	**mean**	**13**	**17**	**2.8**	**471**	**0.05**	**99**	**486**
			95% CL	2.4	10		104		12	186
			CV	27%	63%		30%		16%	52%
	LT	**2003**	**mean**	**nd**	**8.8**	**nd**	**63**	**33**	**nd**	**74**
			95% CL				14			20
			CV				35%			45%
Huron	**LT**	**2001**	**mean**	**13**	**6.9**	**nd**	**352**	**60**	**53**	**445**
			95% CL	2.0	0.56		80	66	14	116
			CV	20%	7%		36%	79%	35%	37%
	LT	**2002**	**mean**	**9.2**	**0.55**	**1.6**	**292**	**16**	**296**	**494**
			95% CL				26		213	134
			CV				14%		104%	39%
	LT	**2003**	**mean**	**9.8**	**nd**	**nd**	**196**	**30**	**25**	**235**
			95% CL				18	13	3.9	21
			CV				15%	31%	21%	15%
Michigan	**LT**	**2001**	**mean**	**24**	**7.1**	**nd**	**560**	**57**	**62**	**683**
			95% CL	5.4	0.8		69	12	23	37
			CV	34%	8%		20%	21%	56%	8%
	LT	**2002**	**mean**	**30**	**12**	**1.8**	**570**	**29**	**64**	**660**
			95% CL	11	8.1		120		50	368
			CV	37%	50%		19%		80%	49%
	LT	**2003**	**mean**	**13.3**	**7.5**	**nd**	**330**	**30**	**32**	**379**
			95% CL	1.7			36	7.7	6.5	42
			CV	20%			17%	18%	30%	18%

Lake	Species	Yr	Statistic	Oxychlordane	Endrin	Mirex	p,p-DDE	p,p-DDD	p,p-DDT	Sum, p,p-DDE+DDT+DDD
Erie	**Walleye**	**2001**	**mean**	**nd**	**nd**	**nd**	**45**	**58**	**nd**	**72**
			95% CL				5.7	44		18
			CV				20%	67%		40%
	Walleye	**2002**	**mean**	**nd**	**nd**	**nd**	**55**	**nd**	**266**	**64**
			95% CL				27		313	14
			CV				66%		104%	25%
	Walleye	**2003**	**mean**	**nd**	**nd**	**nd**	**58**	**32**	**nd**	**86**
			95% CL				14	4.8		20
			CV				39%	17%		40%
Ontario	**LT**	**2001**	**mean**	**13**	**6.8**	**50**	**378**	**32**	**32**	**405**
			95% CL	3.6		20	34		8.6	24
			CV	24%		60%	14%		36%	8%
	LT	**2002**	**mean**	**11**	**nd**	**48**	**522**	**62**	**70**	**627**
			95% CL	1.3		13	160	39	23	212
			CV	9%		41%	47%	79%	51%	52%
	LT	**2003**	**mean**	**9.6**	**nd**	**20**	**241**	**32**	**28**	**276**
			95% CL	0.05		3.6	39	6.5	4.5	48
			CV	0%		29%	26%	21%	22%	29%

Lake	Species	Yr	Statistic	Toxaphene	trans-Nonachlor	cis-Nonachlor	trans-Chlordane	cis-Chlordane
Superior	**LT**	**2001**	**mean**	**414**	**41**	**25**	**4.2**	**12**
			95% CL	55	28	12	1.0	8.8
			CV	12%	60%	44%	20%	66%
	LT	**2002**	**mean**	**1275**	**83**	**48**	**13**	**18**
			95% CL	167	12	12	1.8	3.3
			CV	20%	23%	37%	22%	30%
	LT	**2003**	**mean**	**594**	**25**	**19**	**4.0**	**6.1**
			95% CL	141	12	6.0	1.6	5.1
			CV	38%	74%	51%	61%	44%
Huron	**LT**	**2001**	**mean**	**503**	**78**	**38**	**8.6**	**24**
			95% CL	151	17	9.0	3.3	9.2
			CV	49%	35%	38%	62%	63%
	LT	**2002**	**mean**	**516**	**42**	**23**	**7.6**	**14**
			95% CL	225	4.2	4.3	2.4	3.1
			CV	63%	16%	30%	48%	37%
	LT	**2003**	**mean**	**372**	**41**	**23**	**7.1**	**14**
			95% CL	64	4.8	2.6	2.1	1.6
			CV	28%	19%	18%	48%	19%
Michigan	**LT**	**2001**	**mean**	**579**	**128**	**79**	**23**	**43**
			95% CL	65	29	11	6.4	7.2
			CV	18%	35%	23%	44%	27%
	LT	**2002**	**mean**	**561**	**121**	**67**	**37**	**59**
			95% CL	165	82	29	19	22
			CV	30%	60%	44%	53%	38%
	LT	**2003**	**mean**	**524**	**83**	**45**	**17**	**29**
			95% CL	85	14	5.2	2.6	4.2
			CV	26%	28%	19%	23%	22%

Lake	Species	Yr	Statistic	Toxaphene	trans-Nonachlor	cis-Nonachlor	trans-Chlordane	cis-Chlordane
Erie	**Walleye**	**2001**	**mean**	**127**	**9.8**	**5.7**	**5.4**	**7.0**
			95% CL	24	1.8	1.3	1.0	1.5
			CV	30%	30%	37%	30%	35%
	Walleye	**2002**	**mean**	**94**	**9.0**	**4.4**	**6.1**	**5.4**
			95% CL	22	3.0	1.0	0.53	0.94
			CV	27%	48%	32%	13%	25%
	Walleye	**2003**	**mean**	**231**	**19**	**9.5**	**3.5**	**11**
			95% CL	65	9.6	4.6	1.3	4.9
			CV	45%	83%	77%	56%	69%
Ontario	**LT**	**2001**	**mean**	**238**	**50**	**23**	**13**	**19**
			95% CL	48	15	7.3	4.1	6.9
			CV	32%	48%	50%	52%	58%
	LT	**2002**	**mean**	**202**	**46**	**23**	**13**	**20**
			95% CL	59	9.1	4.4	3.2	2.4
			CV	47%	32%	31%	41%	19%
	LT	**2003**	**mean**	**351**	**39**	**20**	**9.1**	**15**
			95% CL	56	7	3.5	2.1	2.6
			CV	26%	28%	28%	37%	28%

Lake	Species	Yr	Statistic	BDE 47	BDE 66	BDE 99	BDE 100	BDE 153	BDE 154	PBB-153	Hg
Superior	**LT**	**2001**	**mean**	**30**	**1.7**	**12**	**6.4**	**0.55**	**1.6**	**nd**	**141**
			95% CL								27
			CV								31%
	LT	**2002**	**mean**	**44**	**1.7**	**24**	**13**	**1.9**	**1.9**	**1.0**	**293**
			95% CL	13	0.35	9.0	2.9	0.63	0.49	0.69	136
			CV	42%	32%	59%	35%	47%	41%	95%	24%
	LT	**2003**	**mean**	**14**	**1.3**	**12**	**6.8**	**1.5**	**4.8**	**1.0**	**133**
			95% CL	3.2	0.36	4.2	1.9	0.57	1.5	0.32	28
			CV	36%	43%	58%	46%	62%	50%	36%	34%
Huron	**LT**	**2001**	**mean**	**32**	**1.5**	**13**	**9.6**	**2.3**	**4.3**	**2.1**	**148**
			95% CL	13	0.4	6.0	3.4	1.5	2.6	1.2	19
			CV	60%	41%	64%	51%	87%	86%	75%	21%
	LT	**2002**	**mean**	**40**	**1.2**	**8.5**	**8.2**	**2.0**	**8.7**	**14**	**145**
			95% CL	9.2	0.31	2.4	2.2	0.54	7.8	16	49
			CV	37%	37%	46%	44%	37%	137%	143%	17%
	LT	**2003**	**mean**	**27**	**1.2**	**14**	**9.6**	**1.7**	**4.2**	**1.3**	**154**
			95% CL	7.3	0.57	4.1	2.8	1.4	1.7	0.37	12
			CV	44%	68%	47%	47%	122%	67%	45%	13%
Michigan	**LT**	**2001**	**mean**	**108**	**2.3**	**24**	**21**	**3.1**	**7.3**	**1.7**	**160**
			95% CL	20	0.48	8.5	5.0	1.4	3.4	1.4	23
			CV	27%	30%	51%	35%	60%	67%	72%	23%
	LT	**2002**	**mean**	**87**	**4.0**	**32**	**38**	**2.2**	**4.2**	**1.1**	**209**
			95% CL	56.0	1.3	8.5	17	0.71	3.0	0.45	32
			CV	80%	41%	30%	55%	41%	84%	53%	8%
	LT	**2003**	**mean**	**54**	**1.6**	**18**	**19**	**3.3**	**7.3**	**1.2**	**106**
			95% CL	21	0.35	3.9	4.7	0.67	1.6	0.2	14
			CV	60%	33%	33%	38%	31%	33%	22%	21%

Lake	Species	Yr	Statistic	BDE 47	BDE 66	BDE 99	BDE 100	BDE 153	BDE 154	PBB-153	Hg
Erie	**Walleye**	**2001**	**mean**	**5.9**	**nd**	**1.9**	**1.8**	**0.51**	**0.81**	**0.31**	**106**
			95% CL	1.3		0.55	0.64	0.23	0.31	0.015	22
			CV	29%		38%	49%	46%	40%	3%	33%
	Walleye	**2002**	**mean**	**5.4**	**nd**	**0.59**	**0.74**	**18**	**nd**	**nd**	**98**
			95% CL	0.19		0.19	0.21	0.37			52
			CV	4%		37%	29%	1%			27%
	Walleye	**2003**	**mean**	**7.7**	**0.42**	**2.8**	**3.2**	**1.0**	**1.7**	**0.39**	**122**
			95% CL	3.7	0.12	0.89	1.0	0.59	0.56		15
			CV	73%		47%	48%	58%	48%		20%
Ontario	**LT**	**2001**	**mean**	**64**	**1.3**	**9.6**	**10**	**2.2**	**3.8**	**0.95**	**115**
			95% CL	27	0.4	2.7	2.8	1.3	1.9	0.38	10
			CV	62%	42%	42%	43%	90%	77%	46%	14%
	LT	**2002**	**mean**	**48**	**0.73**	**8.6**	**8.3**	**1.7**	**6.5**	**1.3**	**121**
			95% CL	19	0.34	3.4	2.9	1.5	3.9	0.35	46
			CV	59%	66%	60%	53%	126%	85%	37%	20%
	LT	**2003**	**mean**	**28**	**1.1**	**13**	**11**	**3.8**	**7.7**	**1.1**	**108**
			95% CL	9.7	0.15	4.4	3.4	1.4	3.1	0.38	15
			CV	56%	20%	55%	51%	56%	64%	39%	10%

Table 2. Calculated apparent first-order rate constants. Rate constants that are significant at the 95% level are in bold; ++ indicates increasing concentrations. Note that rate constants for Lake Superior – Keweenaw Point are affected by unusually fatty samples in 1991 and 1993 (see Table 1). Calculating lipid-normalized rate constants shows values that are typically consistent with other sites, except the increasing concentrations at the Apostle Islands site.

		Regression performed							Half-life (years)		
Analyte	**Site**	*from*	*to*	**N**	**k (y^{-1})**	**Std Error**	**R^2**	**P**	*upper 95%*		*lower 95%*
PCBs	Michigan - Saugatuck	1973	1982	10	**-0.160**	0.022	0.87	<0.01	6.4	4.3	3.3
	Michigan - Saugatuck	1973	1984	11	**-0.157**	0.017	0.90	<0.01	5.9	4.4	3.5
	Michigan - Saugatuck	1973	1986	12	**-0.165**	0.014	0.93	<0.01	5.2	4.2	3.5
	Michigan - Saugatuck	1974	1982	9	**-0.181**	0.023	0.90	<0.01	5.5	3.8	2.9
	Michigan - Saugatuck	1974	1984	10	**-0.171**	0.018	0.92	<0.01	5.4	4.0	3.3
	Michigan - Saugatuck	1974	1986	11	**-0.176**	0.014	0.95	<0.01	4.8	3.9	3.3
	Michigan - Saugatuck	1975	1982	8	**-0.182**	0.030	0.86	<0.01	6.4	3.8	2.7
	Michigan - Saugatuck	1975	1984	9	**-0.170**	0.022	0.89	<0.01	5.9	4.1	3.1
	Michigan - Saugatuck	1975	1986	10	**-0.177**	0.017	0.93	<0.01	5.0	3.9	3.2
	Superior - Apostle Islands	1977	1982	6	-0.137	0.123	0.24	0.33	++	5.0	1.4
	Superior - Apostle Islands	1977	1984	7	-0.103	0.080	0.25	0.25	++	6.7	2.2
	Superior - Apostle Islands	1977	1986	8	**-0.148**	0.057	0.53	0.04	77	4.7	2.4
	Huron - Rockport	1978	1982	5	-0.070	0.073	0.24	0.41	++	10	2.3
	Huron - Rockport	1978	1984	6	-0.093	0.042	0.54	0.09	++	7.5	3.3
	Huron - Rockport	1978	1986	7	**-0.114**	0.028	0.77	<0.01	17	6.1	3.7
	Michigan - Saugatuck	1982	2002	11	**-0.047**	0.011	0.69	<0.01	29	15	10
	Michigan - Saugatuck	1984	2002	10	**-0.040**	0.012	0.59	<0.01	54	18	10
	Michigan - Saugatuck	1986	2002	9	-0.031	0.013	0.44	0.05	++	22	11
	Superior - Apostle Islands	1982	2002	10	0.021	0.024	0.09	0.40	++	++	20
	Superior - Apostle Islands	1984	2002	9	0.025	0.029	0.09	0.42	++	++	16
	Superior - Apostle Islands	1986	2002	8	0.052	0.030	0.33	0.13	++	++	32
	Huron - Rockport	1982	2002	11	**-0.040**	0.008	0.74	<0.01	32	17	12
	Huron - Rockport	1984	2002	10	**-0.037**	0.009	0.66	<0.01	46	19	12
	Huron - Rockport	1986	2002	9	**-0.031**	0.011	0.53	0.03	138	22	12
	Ontario - Oswego	1982	2002	11	**-0.080**	0.009	0.90	<0.01	11	8.7	7.0
	Ontario - Oswego	1984	2002	10	**-0.073**	0.009	0.88	<0.01	13	10	7.3
	Ontario - Oswego	1986	2002	9	**-0.060**	0.007	0.91	<0.01	16	11	8.9
	Michigan - Sturgeon Bay	1991	2003	7	-0.042	0.018	0.51	0.07	++	17	7.8
	Superior - Keweenaw Point	1991	2003	7	**-0.153**	0.040	0.75	0.01	14	4.5	2.7
	Huron - Port Austin	1991	2003	7	**-0.086**	0.024	0.72	0.02	29	8.1	4.7
	Ontario - N. Hamlin	1991	2003	7	**-0.110**	0.017	0.90	<0.01	10	6.3	4.5
DDTs	Michigan - Saugatuck	1970	1984	14	**-0.137**	0.011	0.93	<0.01	6.1	5.1	4.3
	Michigan - Saugatuck	1970	1986	15	**-0.150**	0.011	0.94	<0.01	5.5	4.6	4.0
	Michigan - Saugatuck	1970	1988	16	**-0.144**	0.009	0.94	<0.01	5.6	4.8	4.2
	Superior - Apostle Islands	1977	1984	7	-0.151	0.062	0.54	0.06	++	4.6	2.2
	Superior - Apostle Islands	1977	1986	8	**-0.201**	0.047	0.75	<0.01	8.1	3.4	2.2
	Superior - Apostle Islands	1977	1988	9	**-0.188**	0.034	0.81	<0.01	6.4	3.7	2.6
	Huron - Rockport	1978	1984	6	-0.116	0.042	0.65	0.05	++	6.0	3.0
	Huron - Rockport	1978	1986	7	**-0.132**	0.027	0.82	<0.01	11	5.2	3.4
	Huron - Rockport	1978	1988	8	**-0.139**	0.019	0.90	<0.01	7.5	5.0	3.7
	Ontario - Oswego	1982	1988	4	-0.127	0.034	0.88	0.06	++	5.4	2.6
	Michigan - Saugatuck	1984	2002	10	**-0.040**	0.012	0.56	0.01	62	17	10
	Michigan - Saugatuck	1986	2002	9	-0.030	0.014	0.40	0.07	++	23	11
	Michigan - Saugatuck	1988	2002	8	**-0.045**	0.014	0.63	0.02	66	16	8.8
	Superior - Apostle Islands	1984	2002	9	0.025	0.030	0.09	0.42	++	++	15
	Superior - Apostle Islands	1986	2002	8	**0.063**	0.023	0.56	0.03	++	++	++
	Superior - Apostle Islands	1988	2002	7	0.064	0.030	0.48	0.09	++	++	53
	Huron - Rockport	1984	2002	10	-0.028	0.013	0.37	0.06	++	24	12
	Huron - Rockport	1986	2002	9	-0.011	0.011	0.13	0.34	++	63	19
	Huron - Rockport	1988	2002	8	-0.002	0.012	0.00	0.90	++	459	23
	Ontario - Oswego	1984	2002	10	**-0.038**	0.010	0.64	<0.01	47	18	11
	Ontario - Oswego	1986	2002	9	**-0.025**	0.009	0.55	0.02	149	27	15
	Ontario - Oswego	1988	2002	8	-0.025	0.011	0.46	0.07	++	27	13
	Michigan - Sturgeon Bay	1985	2003	8	-0.023	0.022	0.15	0.34	++	30	9.0
	Michigan - Sturgeon Bay	1991	2003	7	-0.055	0.029	0.42	0.12	++	13	5.3
	Superior - Keweenaw Point	1985	2003	8	-0.056	0.038	0.26	0.19	++	12	4.6
	Superior - Keweenaw Point	1991	2003	7	**-0.152**	0.017	0.94	<0.01	6.4	4.5	3.5
	Huron - Port Austin	1985	2003	8	**-0.047**	0.015	0.62	0.02	70	15	8.3
	Huron - Port Austin	1991	2003	7	**-0.064**	0.021	0.64	0.03	74	11	5.8
	Ontario - N. Hamlin	1985	2003	8	**-0.057**	0.023	0.51	0.05	962	12	6.1
	Ontario - N. Hamlin	1991	2003	7	**-0.110**	0.018	0.88	<0.01	11	6.3	4.4

Table 2 continued.

Analyte	Site	Regression performed from	to	N	k (y^{-1})	Std Error	R^2	P	Half-life (years) upper 95%		lower 95%
Oxychlordane	Michigan - Saugatuck	1977	1984	7	-0.150	0.076	0.43	0.11	++	4.6	2.0
	Michigan - Saugatuck	1977	1986	8	**-0.158**	0.051	0.61	0.02	21	4.4	2.4
	Michigan - Saugatuck	1977	1988	9	**-0.143**	0.037	0.68	<0.01	13	4.9	3.0
	Superior - Apostle Islands	1977	1986	8	-0.201	0.102	0.39	0.10	++	3.4	1.5
	Superior - Apostle Islands	1977	1988	9	**-0.236**	0.074	0.59	0.02	11	2.9	1.7
	Superior - Apostle Islands	1977	1990	10	**-0.229**	0.055	0.68	<0.01	6.9	3.0	1.9
	Huron - Rockport	1978	1986	6	-0.056	0.045	0.29	0.28	++	12	3.9
	Huron - Rockport	1978	1988	7	**-0.145**	0.048	0.65	0.03	31	4.8	2.6
	Huron - Rockport	1978	1990	8	**-0.121**	0.036	0.66	0.01	20	5.7	3.3
	Michigan - Saugatuck	1984	2002	10	**-0.080**	0.022	0.62	<0.01	24	8.6	5.3
	Michigan - Saugatuck	1986	2002	9	**-0.059**	0.023	0.48	0.04	156	12	6.0
	Michigan - Saugatuck	1988	2002	8	-0.059	0.030	0.39	0.10	++	12	5.2
	Superior - Apostle Islands	1986	2002	8	0.003	0.037	0.00	0.94	++	++	7.9
	Superior - Apostle Islands	1988	2002	7	0.011	0.048	0.01	0.82	++	++	6.2
	Superior - Apostle Islands	1990	2002	6	-0.024	0.060	0.04	0.70	++	28	3.6
	Huron - Rockport	1986	2002	9	-0.045	0.024	0.33	0.11	++	15	6.7
	Huron - Rockport	1988	2002	8	-0.030	0.029	0.15	0.34	++	23	6.8
	Huron - Rockport	1990	2002	7	**-0.072**	0.023	0.67	0.02	51	10	5.3
	Ontario - Oswego	1988	2002	8	0.009	0.020	0.03	0.67	++	++	17
	Ontario - Oswego	1990	2002	7	0.001	0.027	0.00	0.96	++	++	10
	Michigan - Sturgeon Bay	1985	2003	8	**-0.064**	0.018	0.69	0.01	33	11	6.4
	Michigan - Sturgeon Bay	1991	2003	7	-0.064	0.028	0.50	0.07	++	11	5.1
	Superior - Keweenaw Point	1985	1999	6	-0.069	0.077	0.17	0.42	++	10	2.4
	Huron - Port Austin	1985	2003	8	**-0.063**	0.005	0.97	<0.01	13	11	9.3
	Huron - Port Austin	1991	2003	7	**-0.071**	0.005	0.97	<0.01	12	10	8.2
	Ontario - N. Hamlin	1985	2003	8	-0.024	0.016	0.28	0.18	++	28	11
	Ontario - N. Hamlin	1991	2003	7	**-0.053**	0.019	0.61	0.04	181	13	6.8
Dieldrin	Michigan - Saugatuck	1977	2002	16	**-0.072**	0.010	0.77	<0.01	14	10	7.4
	Michigan - Saugatuck	1978	2002	15	**-0.073**	0.012	0.75	<0.01	15	10	7.1
	Michigan - Saugatuck	1979	2002	14	**-0.072**	0.013	0.72	<0.01	16	10	6.9
	Superior - Apostle Islands	1977	2002	15	**-0.025**	0.009	0.39	0.01	110	28	16
	Superior - Apostle Islands	1978	2002	14	**-0.028**	0.009	0.43	0.01	93	25	14
	Huron - Rockport	1978	2002	15	**-0.082**	0.006	0.94	<0.01	10	8.5	7.3
	Ontario - Oswego	1982	2002	11	**-0.072**	0.012	0.80	<0.01	15	10	7.0
	Michigan - Sturgeon Bay	1985	2003	8	-0.023	0.019	0.20	0.27	++	30	10
	Michigan - Sturgeon Bay	1991	2003	7	0.004	0.025	0.00	0.89	++	++	11
	Superior - Keweenaw Point	1985	2003	8	**-0.115**	0.035	0.64	0.02	24	6.0	3.4
	Superior - Keweenaw Point	1991	2003	7	**-0.189**	0.033	0.87	<0.01	6.7	3.7	2.5
	Huron - Port Austin	1985	2003	8	**-0.052**	0.012	0.75	<0.01	31	13	8.5
	Huron - Port Austin	1991	2003	7	-0.042	0.019	0.51	0.07	++	16	7.7
	Ontario - N. Hamlin	1985	2003	8	0.001	0.031	0.00	0.98	++	++	9.3
	Ontario - N. Hamlin	1991	2003	7	0.002	0.050	0.00	0.97	++	++	5.5
Trans Nonachlor	Michigan - Saugatuck	1986	2002	9	**-0.062**	0.025	0.47	0.04	267	11	5.7
	Michigan - Saugatuck	1988	2002	8	-0.064	0.032	0.40	0.09	++	11	4.8
	Michigan - Saugatuck	1990	2002	7	-0.057	0.043	0.26	0.24	++	12	4.2
	Superior - Apostle Islands	1986	2002	8	0.045	0.031	0.26	0.20	++	++	22
	Superior - Apostle Islands	1988	2002	7	0.033	0.040	0.12	0.45	++	++	10
	Superior - Apostle Islands	1990	2002	6	0.016	0.053	0.02	0.78	++	++	5.2
	Huron - Rockport	1986	2002	9	**-0.079**	0.011	0.87	<0.01	13	8.8	6.6
	Huron - Rockport	1988	2002	8	**-0.075**	0.014	0.82	<0.01	17	9.2	6.3
	Huron - Rockport	1990	2002	7	**-0.093**	0.013	0.91	<0.01	12	7.4	5.4
	Ontario - Oswego	1986	2002	9	**-0.077**	0.013	0.84	<0.01	15	9.0	6.4
	Ontario - Oswego	1988	2002	8	**-0.083**	0.016	0.82	<0.01	16	8.4	5.7
	Ontario - Oswego	1990	2002	7	**-0.088**	0.021	0.78	<0.01	20	7.9	4.9
	Michigan - Sturgeon Bay	1985	2003	8	-0.014	0.015	0.12	0.40	++	51	14
	Michigan - Sturgeon Bay	1991	2003	7	-0.031	0.022	0.29	0.21	++	22	8.0
	Superior - Keweenaw Point	1985	2003	8	-0.052	0.054	0.13	0.37	++	13	3.8
	Superior - Keweenaw Point	1991	2003	7	**-0.170**	0.045	0.74	0.01	13	4.1	2.4
	Huron - Port Austin	1985	2003	8	-0.035	0.018	0.40	0.09	++	20	8.9
	Huron - Port Austin	1991	2003	7	**-0.064**	0.022	0.63	0.03	93	11	5.8
	Ontario - N. Hamlin	1985	2003	8	-0.031	0.025	0.20	0.26	++	22	7.5
	Ontario - N. Hamlin	1991	2003	7	**-0.089**	0.017	0.84	<0.01	15	7.8	5.2

Table 2 continued.

		Regression performed							Half-life (years)		
Analyte	**Site**	*from*	*to*	**N**	**k (y^{-1})**	**Std Error**	**R^2**	**P**	*upper 95%*		*lower 95%*
Cis Nonachlor	Michigan - Saugatuck	1986	2002	9	-0.026	0.026	0.13	0.35	++	27	7.9
	Michigan - Saugatuck	1988	2002	8	-0.032	0.033	0.14	0.37	++	22	6.1
	Michigan - Saugatuck	1990	2002	7	-0.023	0.044	0.05	0.63	++	30	5.1
	Superior - Apostle Islands	1986	2002	8	0.045	0.029	0.29	0.17	++	++	27
	Superior - Apostle Islands	1988	2002	7	0.041	0.037	0.19	0.33	++	++	12
	Superior - Apostle Islands	1990	2002	6	0.025	0.051	0.06	0.65	++	++	6.0
	Huron - Rockport	1986	2002	9	**-0.044**	0.014	0.59	0.02	62	16	9.0
	Huron - Rockport	1988	2002	8	**-0.045**	0.018	0.51	0.05	781	16	7.8
	Huron - Rockport	1990	2002	7	-0.056	0.022	0.55	0.06	++	12	6.1
	Ontario - Oswego	1986	2002	9	**-0.057**	0.020	0.54	0.02	71	12	6.7
	Ontario - Oswego	1988	2002	8	-0.061	0.025	0.49	0.05	++	11	5.6
	Ontario - Oswego	1990	2002	7	-0.061	0.034	0.39	0.13	++	11	4.7
	Michigan - Sturgeon Bay	1985	2003	8	0.002	0.019	0.00	0.91	++	++	16
	Michigan - Sturgeon Bay	1991	2003	7	0.005	0.029	0.01	0.87	++	++	10
	Superior - Keweenaw Point	1985	2003	8	-0.052	0.051	0.15	0.35	++	13	3.9
	Superior - Keweenaw Point	1991	2003	7	**-0.159**	0.048	0.68	0.02	20	4.3	2.4
	Huron - Port Austin	1985	2003	8	-0.033	0.017	0.40	0.10	++	21	9.4
	Huron - Port Austin	1991	2003	7	**-0.088**	0.026	0.69	0.02	34	7.9	4.5
	Ontario - N. Hamlin	1985	2003	8	-0.042	0.019	0.44	0.07	++	16	7.7
	Ontario - N. Hamlin	1991	2003	7	**-0.086**	0.016	0.86	<0.01	15	8.1	5.5
Trans Chlordane	Michigan - Sturgeon Bay	1991	2003	5	0.016	0.033	0.08	0.65	++	++	7.9
	Michigan - Saugatuck	1986	2002	6	-0.028	0.016	0.44	0.15	++	25	10
	Superior - Keweenaw Point	1991	2003	5	**-0.108**	0.011	0.97	<0.01	10	6.4	4.8
	Huron - Port Austin	1991	2003	5	-0.049	0.016	0.76	0.06	++	14	6.9
	Ontario - N. Hamlin	1991	2003	5	-0.049	0.036	0.38	0.27	++	14	4.2
	Ontario - Oswego	1986	2002	5	-0.006	0.025	0.02	0.81	++	110	8.2
Cis Chlordane	Michigan - Saugatuck	1986	2002	9	-0.074	0.035	0.39	0.07	++	9.4	4.4
	Michigan - Saugatuck	1988	2002	8	-0.081	0.045	0.35	0.12	++	8.6	3.6
	Michigan - Saugatuck	1990	2002	7	-0.070	0.059	0.22	0.29	++	10	3.1
	Superior - Apostle Islands	1986	2002	8	0.033	0.038	0.11	0.42	++	++	12
	Superior - Apostle Islands	1988	2002	7	0.011	0.047	0.01	0.83	++	++	6.4
	Superior - Apostle Islands	1990	2002	6	0.012	0.065	0.01	0.87	++	++	4.1
	Huron - Rockport	1986	2002	9	**-0.081**	0.021	0.67	<0.01	23	8.6	5.3
	Huron - Rockport	1988	2002	8	**-0.087**	0.027	0.63	0.02	34	8.0	4.5
	Huron - Rockport	1990	2002	7	-0.058	0.029	0.45	0.10	++	12	5.3
	Ontario - Oswego	1986	2002	9	**-0.090**	0.022	0.71	<0.01	18	7.7	4.9
	Ontario - Oswego	1988	2002	8	**-0.070**	0.024	0.59	0.03	58	10	5.4
	Ontario - Oswego	1990	2002	7	-0.076	0.032	0.54	0.06	++	9.1	4.4
	Michigan - Sturgeon Bay	1985	2003	8	**-0.044**	0.015	0.60	0.02	83	16	8.6
	Michigan - Sturgeon Bay	1991	2003	7	-0.050	0.023	0.48	0.09	++	14	6.3
	Superior - Keweenaw Point	1985	2003	8	-0.094	0.046	0.41	0.09	++	7.4	3.4
	Superior - Keweenaw Point	1991	2003	7	**-0.186**	0.046	0.77	<0.01	10	3.7	2.3
	Huron - Port Austin	1985	2003	8	**-0.077**	0.017	0.77	<0.01	20	9.1	5.9
	Huron - Port Austin	1991	2003	7	**-0.088**	0.026	0.69	0.02	34	7.9	4.5
	Ontario - N. Hamlin	1985	2003	8	-0.042	0.019	0.44	0.07	++	16	7.7
	Ontario - N. Hamlin	1991	2003	7	**-0.109**	0.028	0.75	0.01	19	6.4	3.8
Toxaphene	Michigan - Saugatuck	1986	2002	9	-0.093	0.043	0.40	0.07	++	7.4	3.6
	Michigan - Saugatuck	1988	2002	8	-0.098	0.055	0.35	0.12	++	7.1	3.0
	Michigan - Saugatuck	1990	2002	7	-0.084	0.073	0.21	0.30	++	8.2	2.5
	Superior - Apostle Islands	1988	2002	7	-0.023	0.051	0.04	0.67	++	30	4.5
	Superior - Apostle Islands	1990	2002	6	-0.034	0.070	0.05	0.66	++	20	3.0
	Huron - Rockport	1986	2002	9	-0.048	0.022	0.40	0.07	++	14	6.9
	Huron - Rockport	1988	2002	8	-0.028	0.025	0.18	0.29	++	24	7.8
	Huron - Rockport	1990	2002	7	-0.031	0.033	0.15	0.38	++	22	6.0
	Ontario - Oswego	1992	2002	6	-0.083	0.081	0.21	0.36	++	8.3	2.2
	Michigan - Sturgeon Bay	1991	2003	7	-0.062	0.051	0.23	0.27	++	11	3.6
	Superior - Keweenaw Point	1991	2003	7	**-0.205**	0.048	0.78	<0.01	8.6	3.4	2.1
	Huron - Port Austin	1991	2003	7	**-0.118**	0.028	0.77	<0.01	16	5.9	3.6
	Ontario - N. Hamlin	1991	2003	7	-0.118	0.071	0.36	0.16	++	5.9	2.3
HCB	Michigan - Saugatuck	1994	2002	5	0.002	0.052	0.00	0.98	++	++	4.3
	Michigan - Sturgeon Bay	1995	2003	5	-0.089	0.060	0.42	0.24	++	7.8	2.5
	Superior - Keweenaw Point	1995	2003	5	-0.090	0.049	0.53	0.16	++	7.7	2.8
	Huron - Port Austin	1995	2003	5	-0.090	0.061	0.42	0.24	++	7.7	2.5
	Ontario - N. Hamlin	1995	2003	5	-0.101	0.059	0.50	0.18	++	6.8	2.4
	Ontario - Oswego	1994	2002	5	-0.036	0.046	0.17	0.49	++	19	3.8

Table 2 continued.

Analyte	Site	Regression performed from	to	N	k (y^{-1})	Std Error	R^2	P	Half-life (years) upper 95%		lower 95%
Heptachlor epoxide B	Michigan - Saugatuck	1992	2002	6	-0.073	0.043	0.42	0.16	++	10	3.6
	Michigan - Sturgeon Bay	1985	2003	8	**-0.049**	0.004	0.96	<0.01	18	14	12
	Michigan - Sturgeon Bay	1991	2003	7	**-0.044**	0.006	0.92	<0.01	24	16	12
	Superior - Apostle Islands	1992	2002	5	-0.119	0.103	0.31	0.33	++	5.8	1.6
	Superior - Keweenaw Point	1985	1999	5	-0.004	0.049	0.00	0.95	++	192	4.4
	Huron - Port Austin	1985	2003	8	-0.053	0.029	0.37	0.11	++	13	5.6
	Huron - Port Austin	1991	2003	7	-0.051	0.046	0.20	0.32	++	14	4.1
	Huron - Rockport	1992	2000	5	-0.089	0.046	0.55	0.15	++	7.8	2.9
	Ontario - N. Hamlin	1985	1999	6	-0.002	0.025	0.00	0.94	++	356	10
	Ontario - Oswego	1992	2000	5	-0.037	0.018	0.58	0.13	++	19	7.4
Mirex	Huron - Rockport	1992	2002	5	-0.153	0.084	0.53	0.17	++	4.5	1.6
	Ontario - N. Hamlin	1991	2003	7	**-0.238**	0.025	0.95	<0.01	4.0	2.9	2.3
	Ontario - N. Hamlin	1993	2003	6	**-0.264**	0.028	0.96	<0.01	3.7	2.6	2.0
	Ontario - Oswego	1992	2002	6	**-0.161**	0.054	0.69	0.04	62	4.3	2.2
	Ontario - Oswego	1994	2002	5	-0.183	0.080	0.64	0.11	++	3.8	1.6
OCS	Huron - Port Austin	1995	2003	5	0.045	0.017	0.69	0.08	++	++	67
	Huron - Rockport	1994	2002	5	-0.006	0.017	0.04	0.73	++	108	11
	Michigan - Saugatuck	1994	2002	5	-0.008	0.069	0.00	0.92	++	91	3.1
	Michigan - Sturgeon Bay	1995	2003	5	0.105	0.050	0.59	0.13	++	++	13
	Ontario - N. Hamlin	1995	2003	5	-0.012	0.021	0.10	0.61	++	58	8.8
	Ontario - Oswego	1994	2002	5	-0.121	0.058	0.59	0.13	++	5.7	2.3
Endrin	Michigan - Saugatuck	1994	2002	5	-0.080	0.088	0.21	0.43	++	8.7	1.9
	Michigan - Sturgeon Bay	1995	2003	5	-0.001	0.027	0.00	0.97	++	568	8.1
	Huron - Rockport	1994	2002	5	-0.306	0.155	0.57	0.14	++	2.3	0.9

Table 3. Ranges of observed half-lives of selected contaminants in lake trout from the Great Lakes, taken from Table 2. “Early” indicates half-lives from 1970s to mid-80s; “late” indicates those from mid-1980s forward. Values in italics without bolding are not statistically significant (no longer changing; infinite half-life). Increasing concentrations (typically not significant) are indicated with an arrow.

	Superior Apostle Islands		**Huron Rockport**		**Michigan Saugatuck**		**Ontario Oswego**	
	early	**late**	**early**	**late**	**early**	**late**	**early**	**late**
PCBs	**5**-*7*	↑	**6**-*10*	*17-22*	**4**	**15**-*22*		**10**
DDT	**3**-*5*	↑	**5**-*6*	*24->30*	**5**	**16**-*23*	*5*	**18**-*27*
Oxychlordane	**3**	↑-*28*	**5**-*12*	**10**-*23*	**4-5**	**9**-*12*		↑
Dieldrin	**25-28**		**9**		**10**		**10**	
t-Nonachlor		↑		**7-9**		**11**-*12*		**8-9**
c-Nonachlor		↑		*12*-**16**		*22-30*		*11*-**12**

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